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(54) **Method for reducing engine friction**

(57) A method for reducing friction in the operation of an internal combustion engine comprising delivering to the internal combustion engine a fuel comprising gasoline and a friction-reducing additive based on a N. N-bis(hydroxyalkyl)-alkylamine.

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Description

This invention relates to methods for using friction reduction agents in engine fuels. In particular, this invention relates to methods for reducing friction in the operation of an internal combustion engine using ethoxylated amines.

A major concem today is finding methods to reduce engine friction and fuel consumption in internal combustion engines which are safe for the environment and economically attractive. One means is to treat moving parts of such engines with lubricants containing friction reducing additives. Considerable work has been done in this area.

U.S Patent No. 4,086,172 (Lowe) discloses a lubricating oil additive composition which is stated to impart improved oxidation properties to crankcase lubricants comprising an antioxidant and certain oil-soluble hydroxy amines. Among the compounds disclosed is "ETHOMEEN 18-12™" which has the formula $C_{18}H_{37}N\text{-}(CH_2CH_2OH)_2$.

U.S. Patent No. 4,129,508 (Friihauf) discloses a lubricant and fuel composition comprising one or more reaction products of a hydrocarbon-substituted succinic acid or anhydride with one or more polyalkylene glycols or monoethers thereof, one or more organic basic metal salts, and one or more alkoxylated amines. The alkoxylated amines include commercially available ETHOMEEN™ products, including ETHOMEEN C/12™. Friihauf states that lubricant and fuel compositions containing these additives possess improved demulsifier properties.

U.S. Patent No. 4,231,883 (Malec) discloses lubricating oils and engine fuels containing an hydroxyalkyl amine compounds as additives. An example of such an additive is N,N-bis(2-hydroxyethyl)-n-dodecylamine. It is stated that lubricating oils formulated for the crankcase of an internal combustion engine and comprising these additives reduce engine friction and improve fuel mileage. It is further stated that a useful concentration of the additive in the crankcase oil is 0.05 to 3 weight percent.

U.S. Patent No. 4,409,000 (LeSuer), now Re. 32,174, discloses a combination of hydroxy amines and hydrocarbon-soluble carboxylic dispersants as engine and carburetor detergents for liquid fuels. One of the preferred hydroxy amines is ETHOMEEN C/12™. It is disclosed that such additives inhibit the formation of unwanted deposits in the carburetor and engine.

U.S. Patent No. 4,836,829 (Zimmerman) discloses a gasoline additive comprising a mixture of certain hydroxy amines and certain demulsifying agents that are the to be useful for reducing and/or preventing fouling in a multi-port, electronically controlled fuel injection system for an intemal combustion engine. Preferred additive mixtures also include an amine oxide as a second anti-fouling agent. A preferred hydroxy amine is bis(2-hydroxy ethyl) cocoamine. The concentration of the antifouling agent in the fuel is typically from 2 to 200 ppm, preferably 40 to 120 ppm, based on the total weight of the fuel composition.

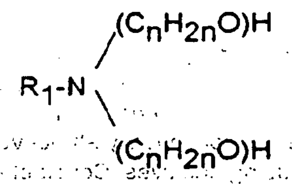
As indicated above, friction reducing additives are generally added to lubricating fuel oils, not engine fuels. We have found, however, that it is possible to reduce engine friction by delivering a friction reducing agent to the engine in the form of a friction reducing additive in the gasoline used to fuel the engine. By doing so, increased power and fuel economy may be obtained.

The present invention provides a method for reducing friction in the operation of an internal combustion engine by delivering to the internal combustion engine a fuel comprising gasoline and a friction-reducing additive. The friction-reducing additive comprises a mixture of an effective amount of N,N-bis(hydroxyalkyl)alkylamine and at least one detergent. The detergent preferably is a polyalkeneamines, a polyetheramine or a Mannich base-type condensation products.

In order to reduce engine friction, the gasoline preferably comprises at least certain minimum amounts of N,N-bis (hydroxyalkyl)-alkylamine, that being at least 7 g.hl.$^{-1}$ (grams per hectoliter) (at least 25 ptb - pounds per thousand barrels) of gasoline. The total amount of N,N-bis(hydroxyalkyl)-alkylamine and detergent should preferably be at least 28.5 g.hl.$^{-1}$ (100 ptb). Gasoline fuels containing less than these minimum required amounts of the above mentioned ingredients do not adequately reduce engine friction and/or improve gasoline mileage. The preferred gasolines comprise a mixture of at least 11.4 g.hl.$^{-1}$ (40 ptb) of N,N-bis(hydroxyethyl)-dodecylamine as the effective friction reducing agent and at least 28.5 g.hl.$^{-1}$ (100 ptb) of detergent.

Detailed Description

The N,N-bis(hydroxyalkyl)-alkylamine typically has short chain (C_2-C_4) hydroxyalkyl groups and a long chain (C_8-C_{18}) alkyl group. A preferred compound of this type is bis(2-hydroxy ethyl)cocoamine (also known as bis-ethoxylated cocoamine). The compounds used as the effective friction reducing agents have the following structure:



in which R_1 is a hydrocarbyl group having from 8 to 18 carbon atoms and n is an integer from 2 to 4. The preferred N, N-bis(hydroxyethyl) n-dodecylamine is usually derived from coconut fatty acid so that the R_1 substituent generally ranges from C_8 to C_{18}, with C_{12} and C_{14} groups predominanting, mostly straight chain.

The preferred friction reducing agent is available commercially as ETHOMEEN C/12™ from Akzo Nobel, of Chicago, Illinois. Alternatively, it may be prepared using any of the methods for preparing ethoxylated amines which are well known in the art. Generally, ethoxylated amines may be prepared by the reaction of the appropriate hydrocarbyl amine with ethylene oxide. often catalyzed, to form the corresponding ethoxylated amine.

The concentration of the hydroxyalkyl alkylamine (HAAA) additive in the gasoline is usually at least 7 g.hl.$^{-1}$(25 ptd), preferably at least 11.4 g.hl.$^{-1}$(40 ptb), more preferably at least 14.25 g. hl.$^{-1}$(50 ptb), and even more preferably at least 17.1 g. hl.$^{-1}$ (60 ptb). More than 28.5 g. hl.$^{-1}$(100 ptb) is usually not required for effective friction reduction.

The other preferred component of the fuel additive is at least one detergent which is typically a polyalkeneamine, a polyetheramine or a Mannich base, all of which are conventional materials. Generally, these detergents reduce and/or prevent deposits which have a tendency to form in carburetors, fuel injectors, intake valves, and intake ports, thereby improving engine performance. Such detergent agents also improve fuel economy and reduce internal combustion engine exhaust emissions.

The preferred polyalkeneamine detergents are selected from the group consisting of polymeric 1-amines, including polyisobutylene-amine. High vinylic content polyisobutylene-amines are most preferred. Suitable polyisobutylene-amines are described in U.S patent 5,004,478 (Vogel et al.), 5,112,364 (Rath et al.), and DE 3942860. Preferred polyisobutylene-amines have a weight average molecular weight of 500 to 2000.

Such polyalkeneamines are available from normal commercial sources or may be prepared by the amination of high vinylic content polyolefins having a weight average molecular weight of from 500 to 3,000 or greater, using conventional methods. Polyisobutylene amines are generally prepared by chlorination or hydroformylation of reactive polyisobutylene and subsequent amination with ammonia, hydrocarbyl amine, hydrocarbyl diamine, hydrocarbyl polyamine, alkoxylated hydrocarbyl amines, or mixtures thereof. Ammonia, ethylenediamine, diethylenetriamine, triethylenetetramine, tetraethylenepentamine, piperazines, hexamethylenediamine, hydroxyalkyl ethylenediamines, hydroxyalkyl triethylenetetraamines, and the like can be incorporated into the polyalkeneamines. Such amines can be prepared by the chlorination or halogenation of appropriate polymeric olefins, and subsequently converted into corresponding polyalkene derivatives using these or other known methods of manufacture.

Alternative preferred detergent agents are the Mannich base condensation products. The Mannich base detergents are condensation products of hydrocarbyl phenols, aldehyes, and amines. In the manufacture of the Mannich base reaction products, formaldehyde (or another aldehyde) is condensed with ammonia (or with salts of primary or secondary amines, or with amides) and a compound containing an active hydrogen. For example, the Mannich base reaction of hydrocarbyl phenols, aldehydes, and amines are generally prepared by the alkylation of phenol or phenolics with hydrocarbyl groups having from 10 to 150 or more carbon atoms. For instance, long chain olefins or polymeric olefins such as polypropylene, polyisobutylene, or mixed polymerized or oligomerized olefins can be used in the phenol alkylation step. The substituted phenol is then reacted with a carbonyl source and an amine. Carbonyl sources include aldehydes such as formaldehyde, acetaldehyde, propanal, butanal, 2-ethylhexanal and the like. In addition aromatic aldehydes may be used to provide a carbonyl source. For instance, benzaldehyde, vanillin, salicylaldehyde and cinnamaldehyde may be used. Dicarbonyl compounds such as glyoxyls as well as carbonyl generating sources such as paraformaldehyde can also be used in some aspects of the invention.

Amines useful in preparation of the Mannich base condensation product include primary or secondary amines. Fatty amines, hydroxyl-containing amines, polyamines such di-, tri-, tetra-, and pentamines or polymeric amines can be used in some aspects of this invention. For example, linear and cyclic C_2-C_6 alkylene di-, tri-, tetra, and pentamines, polyamines, and their substituted polyfunctional derivatives can be used. Subtituted derivatives, as used herein, refer to substitution with substituents such as halo, hydroxy, alkoxy, nitro, thio, carbalkoxy and alklythio substituents.

Amines suitable for use in the Mannich base reaction also include ethylene diamines, diethylene triamines, triethylene tetramines, tetraethyleneamine pentamines, propylene diamines, propylene triamines, ethylene triamines, ethylene tetramines, ethylene pentamines, hexamethylene diamines, and the like. Amines containing from 2 to 30 or more carbon atoms are preferred

The Mannich base product can also react further in at least three additional ways If it is a primary or secondary

amine, it may condense with one or two additional molecules of aldehyde and active compound to yield a product such as $N(CH_2CH_2COR)_3$ or the like. If the active hydrogen compound has two or three active hydrogens, the Mannich base may condense with one or two additional molecules of aldehyde and ammonia or amine to yield a product such as $(H_2NCH_2)_3$ CCOR or the like. Alternatively, another further reaction consists of condensation of the Mannich base with excess formaldehyde yielding a product such as $H_2C{=}NCH_2CH_2COR$ or the like.

The polyetheramines may be used instead of the polyamines decribed above but generally they are less preferred. Typical polyether primary amines include those of the formula R17-O(C2H3(R18)O)[n]C3H6NH[2] where R17 is an alkyl-substituted phenyl group containing 14 to 26 carbon atoms, C6 to C30 alkyl group or C7 to C30 aralkyl group, n is an integer ranging from 2 to 10 and R18 is independently hydrogen or methyl. These alkyl-substituted phenol-derived polyetheramines are sold by Texaco Chemical Co. under the trademark Surfonamine. Those available commercially include Surfonamine MNPA-380™ (nonylphenyl-1EO-2PO-NH2), Surfonamine MNPA-510™ (nonylphenyl-4EO-2PO-NH2), Surfonamine MNPA-750™ (nonylphenyl-9.5EO-2PO-NH2) and Surfonamine MNPA-860™ (nonylphenyl-12EO-2PO-NH2). See, for example, US 5509943. Other polyetheramines are described in U.S. 5,112,364 (gasoline-engine fuels which contain small amounts of a polyetheramine is prepared by reductive amination of a phenol-initiated or alkylphenol-initiated polyether alcohol with ammonia or a primary amine) and European Patent Application Publication No. 310 875 (fuels for spark ignition engines containing a polyetheramine additive prepared by first propoxylating and/or butoxylating an alkanol or primary or secondary alkylmonoamine and then aminating the resulting polyether with ammonia or a primary aliphatic amine).

The present invention can be used with gasoline fuels intended for use in spark ignition internal combustion engines. Such gasoline fuels include oxygenated gasolines, reformulated gasolines and gasohols. In order to achieve friction reduction and improved fuel economy, the total amount of HAAA, e.g., ETHOMEEN C/12™ and detergent is preferably is at least 26.5 $g.hl.^{-1}$ (100 ptb), more preferably at least 42.75 $g.hl.^{-1}$(150 ptb), and even more preferably between 48.45 to 57 $g.hl.^{-1}$(170 to 200 ptb). Typically, the total amount of HAAA plus detergent ranges from 26.5 to 85.5 $g.hl.^{-1}$ (100 to 300 ptb), preferably between 40 to 57 $g.hl.^{-1}$(140 to 200 ptb). The amount of detergent is typically from 2.85 to 5.7 $g.hl.^{-1}$(10 to 200 ptb, and usually 20 to 42.75 $g.hl.^{-1}$(70 to 150 ptb).

An excess of HAAA or detergent can be used. for instance, the ratio of detergent to HAAA can vary from 10:1 to 1:10, with ratios of from 5:1 to 1:5 being preferred. In other preferred embodiments of the present invention the ratio of detergent to HAAA ranges from 1:1 to 5:1. In some more preferred embodiments of the present invention the ratio of detergent to HAAA is 2-3:1.

Additional components such as corrosion inhibitors, demulsifiers, solubilizers, carrier fluids, and the like can also be used with this invention. For example, additives which are useful in practicing the method of this invention further comprise at least one demulsifier selected from the group consisting of acylated polyglycols; alkyl aryl sulfonates; polyglycols; fatty acid alkylamine reaction products; oxyalkylated alkylphenol (formaldehyde) resins; and oxyalkylated alkylphenolic (formaldehyde) resins and polyoxyalkylene glycol. A preferred demulsifier is oxyalkylated alkylphenolic (formaldehyde) resins, with or without polyoxyalkylene glycol. Suitable demulsifiers which are available commercially include the following materials: Tolad T-284™ (solution of acylated polyglycols in aromatic hydrocarbons), Tolad T-286™ (alkyl aryl sulfonates, polyglycols, oxyalkylated alkylphenol-formaldehyde resins in aromatic hydrocarbons and isopropyl alcohol), Tolad T-326™ (oxyalkylated alkylphenol-formaldehyde resins and polyglycols in aromatic naphtha), Tolad T-500 ™(oxyalkylated alkylphenol-formaldehyde resins in aromatic hydrocarbons and alkanols), Tolad T-292™ (oxyalkylated alkylphenol formaldehyde resins in aromatic hydrocarbons and isopropanol), Tolad T-347™ (oxyalkylated alkylphenol formaldehyde resins and acylated polyglycols in aromatic hydrocarbons and methanol), Tolad T-370™ (polyglycols in aromatic hydrocarbons), Nalco 5450™ (hydrocarbon blend of alkylphenol formaldehyde resin polyoxyalkylene polyether), Nalco 5451™ (polyglycolated polyol esters and polyglycolated alkylphenol/formaldehyde resin in aromatic solvent), Nalco 5452™ (polyethers and oxyalkylated alkylphenol/formaldehyde resin adducts in aromatic solvent), Nalco 5453™(oxyalkylated alkylphenol/formaldehyde resin adducts in aromatic solvent), Nalco 85BD-194™ (ethoxylated nonyl phenol/formaldehyde resin in hydrocarbon solvent), Nalco 5455™ (oxyalkylated alkyl phenol-formaldehyde resin in aromatic solvent). (Tolad is a trademark of Tretolite Division of Petrolite Corporation, St. Louis, Missouri). Nalco is a trademark of Nalco Chemical Company, Oak Brook, Illinois.

Suitable carrier fluids include propylene glycol-based polyethers and butylene glycol-based polyethers, aromatic and aliphatic esters, polyalkylenes, and mineral oils.

The following examples illustrate the invention.

Example 1

An equal weight mixture of bis(2-hydroxyethyl)cocoamine and 900 MW polyisobutylene amine (primarily amine derived from terminal polymeric vinylic groups) was prepared by blending at approximately 75° C. This concentrate was used for testing and can be used to prepare fuel blends by appropriate dilution

Example 2

A gasoline blend suitable for use in internal combustion engines is prepared by admixing approximately 14.25 g. $hl.^{-1}$ (50 ptb) of bis(2-hydroxyethyl) cocoamine and approximately 20 g. $hl.^{-1}$ (70 ptb) of a polymeric 900 MW polyisobutyleneamine (consisting of primarily more than 70% of terminal amine groups.)

Example 3

An equal mixture of bis(2-hydroxyethyl)cocoamine and a Mannich base reaction product of polyisobutenyl (approximately 900 MW) alkylated phenol, formaldehyde, and triethylene tetramine based ethylene amines was prepared by blending the two components at approximately 75° C. This concentrate was used for testing and can be used to prepare fuel blends by appropriate dilution.

Example 4

A gasoline blend suitable for use in an internal combustion engine is prepared by admixing approximately 14.25 g. $hl.^{-1}$ (50 ptb) of bis(2-hydroxyethyl)cocoamine and approximately 20 g. $hl.^{-1}$ (70 ptb) of a Mannich base reaction product of polyisobutenyl (approximately 900 MW) alkylated phenol, formaldehyde, and triethylene tetramine based ethylene amines.

Comparative Example A

A carboxylate dispersant was prepared according to the generalized teaching of U.S. patent No. 4,409,000. A polyisobutylene (having a MW of 900) was reacted with maleic anhydride to form a polyisobutenyl succinic anhydride. This hydrocarbyl succinic anhydride was then reacted with tetraethylene pentamine to form a polyisobutenyl succinimide detergent.

An equal weight mixture of bis(2-hydroxyethyl)cocoamine and succinimide detergent was prepared by blending at approximately 75° C.

High Temperature Performance Cleanliness

The products of Examples 1, 3, and Comparative Example A were evaluated with respect to cleanliness during thermal decomposition using thermogravimetric analysis as shown in Table 1 below. Thermogravimetric analysis was performed by heating the sample at 20° C/min in air flowing at 100 ml/min using a TA thermogravimetric analyzer. The percent residue remaining at 325° C, 425° C, and 525° C was recorded. Little or no residue is most desirable.

Example	% Residue at 325° C	% Residue at 425° C	% Residue at 525° C
1	4.3	1.5	0.5
3	9.4	2.9	0.9
Comparative Example A	35.5	6.3	2.4

As can be seen from the thermogravimetric analyses results, the products of this invention show exceptional cleanliness and high temperature decomposition, when compared to Comparative Example A at a various high temperatures.

The following data demonstrate that the use of Ethomeen C/12™ results in friction reduction.
Ethomeen C/12™ was added to a 5W-30 engine oil which was usd as the crakcase oil in a motored (non-fuelled) 1.51 Toyota engine. The results are shown below.

Lube spiking results	
Ethomeen C/12™ in a 5W-30; 1.5L Toyota - Motored	
wt% (Spiked in lube)	% benefit (decrease in Motoring Torque)
0.25	3.3
0.50	14.0
Speed: 1500 rpm	
Temperature: 225°F (107°C)	

The lube spiking test was repeated at various concentration levels in a 3.81 Buick fired (fuelled) engine, with the results shown below.

Lube spiking results	
Ethomeen C/12™ in a 5W-30; 3.8L Buick-Fired	
wt% (spiked in lube)	% benefit (decrease in BSFC)
0	0.0
0.1	2.0
0.2	2.4
0.3	2.4
0.4	3.5
0.5	4.4
0.6	6.1
Speed: 1500 rpm Load: 6.6 HP Temperature: 275°F (135°C)	

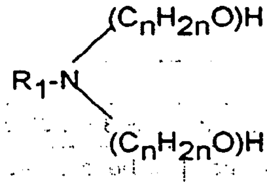
The effectiveness of the hydroxyamine as a fuel additive for reducing engine friction is shown by the results below which show the decrease in BSFC when 50 ptb of Ethomeen C/12™ is used as a gasoline additive for the gasoline fuel fed to a 3.8 l Buick fired engine on 5W-30 engine oil.

Fueled engine test results	
Effect of Ethomeen C/12™ as gasoline additive (50 ptb)	
hours (of engine operation)	% benefit (decrease in BSFC)
0	0.0
32	2.0
64	2.3
96	2.3
128	4.2
160	5.0
192	6.0
224	6.1
256	6.1
288	6.1
BSFC = brake specific fuel consumption (Fuel economy evaluations on a stationary engine stand) Speed: 1500 rpm Load: 6.6 HP Temperature: 275°F (135°C)	

Claims

1. A method for reducing friction in the operation of an internal combustion engine which comprises delivering to the internal combustion engine a fuel comprising gasoline and friction-reducing additive comprising an N,N-bis(hydroxyalkyl)alkylamine.

2. The method of claim 1 in which the bis-(hydroxyalkyl)alkylamine has the formula:



in which R_1 is a hydrocarbyl group having from 8 to 18 carbon atoms and n is an integer from 2 to 4.

3. The method of claim 2 in which the bis(hydroxyalkyl)alkylamine is bis(hydroxyethyl) dodecylamine.

4. The method of any of claims 1 to 3 in which the gasoline includes at least 11.4 $g.hl^{-1}$ (40 ptb) N,N-bis(hydroxyethyl)-dodecylamine, preferably 14.25 to 28.5 $g.hl^{-1}$ (50 to 100 ptb) N,N-bis(hydroxyethyl)-dodecylamine.

5. The method of any preceding claim in which the gasoline includes a detergent.

6. The method of claim 5 in which the detergent is a polyalkeneamine, a polyetheramine or a Mannich base reaction product.

7. A method according to claim 5 or claim 6 in which the weight ratio of detergent to N,N-bis(hydroxyalkyl) alkylamine is in the range 1:1 to 5:1.

8. A method according to claim 1 in which the gasoline comprises at least 28.5 $g.hl^{-1}$ (100 ptb) of a friction-reducing additive combination which comprises N,N-bis(hydroxyethyl)dodecylamine and a detergent, in which the ratio of detergent to N,N-bis(hydroxyethyl)dodecylamine is between 1:1 and 5:1.

9. The use in a motor gasoline fuel of an additive for reducing friction in the engine burning the fuel of a friction-reducing additive comprising an N,N-bis(hydroxyalkyl)alkylamine.

10. Use according to claim 9 in which the N,N-bis(hydroxyalkyl)alkylamine is present in the fuel in an amount of at least 7 $g.hl^{-1}$ (25 ptb).

11. A motor gasoline fuel comprising a fuel base and a friction reducing additive comprising an N,N-bis(hydroxyalkyl) alkylamine and, optionally, a detergent.

12. A fuel according to claim 11 in which the friction reducing additive is included in an amount of at least 28.5 $g.hl^{-1}$ (100 ptb) and in which the ratio of detergent to N,N-bis(hydroxyalkyl)alkylamine is in the range 1:1 to 5:1.

13. A fuel according to claim 11 or claim 12 in which the detergent is a polyalkeneamine, a polyetheramine or a Mannich base reaction product.

European Patent Office

EUROPEAN SEARCH REPORT

Application Number
EP 98 30 2599

DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.Cl.6)
D,X	US 4 231 883 A (MALEC) 4 November 1980 * column 1, line 26 - line 27 * * column 3, line 61 - line 68 *	1-3,5,6,9,11,13	C10L1/22 C10L10/04
P,X	EP 0 780 460 A (EXXON) 25 June 1997 * the whole document *	1-4,9-11	
D,X	US 4 129 508 A (FRIIHAUF) 12 December 1978 * column 9 - column 10 *	11-13	
X	WO 92 14805 A (LUBRIZOL) 3 September 1992 * page 31; claim 1 * * page 34 *	11	
D,X	FR 2 518 114 A (LUBRIZOL) 17 June 1983 * page 19 *	11,12	
X	US 3 115 400 A (MARSH ET AL.) 24 December 1963 * the whole document *	11	TECHNICAL FIELDS SEARCHED (Int.Cl.6)
D,X	EP 0 237 356 A (EXXON) 16 September 1987 * claim 8 *	11	C10L
A	WO 96 21709 A (EXXON) 18 July 1996 * the whole document *	1-13	

The present search report has been drawn up for all claims

Place of search	Date of completion of the search	Examiner
THE HAGUE	16 July 1998	De La Morinerie, B

CATEGORY OF CITED DOCUMENTS

X : particularly relevant if taken alone
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A : technological background
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E : earlier patent document, but published on, or after the filing date
D : document cited in the application
L : document cited for other reasons
& : member of the same patent family, corresponding document

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